

1S 4313539

(s) at one time; forming the upper electrode; and conducting heat-treatment in an oxygen atmosphere at about 550.degree. C. for about 60 minutes.

(91) The crystallizing heat-treatment may be conducted either as batch-crystallization for collectively heat-treating the film deposited to a desired thickness by repeating the applying and drying steps several times, or as a crystallization process for heat-treating the films on a film-by-film basis after every applying and drying step. Moreover, the time for the heat-treatment is not specifically limited. The heat-treatment may be conducted for several minutes to several hours in a diffusion furnace, or may be conducted for several tens of seconds to several minutes by RTA (Rapid Thermal Annealing). In short, the time for the heat-treatment is determined so that sufficient crystallization can be achieved in respective heat-treatment processes.

(92) In general, heat-treatment for crystallizing an oxide must be conducted in an oxygen atmosphere due to its requirement to supply a sufficient amount of oxygen. On the other hand, in the methods such as a sol-gel method and an MOD method, a large amount of oxygen is contained in the raw materials such as metal alkoxide and salt. Therefore, crystallization can be conducted even in an inert atmosphere such as nitrogen or argon. However, in order to improve the ferroelectric characteristics and suppress a leakage current, it is preferable to conduct heat-treatment at about 400.degree. C. to about 650.degree. C. after the formation of the upper electrode.

(93) According to the MOCVD method, the ferroelectric film is formed as

L Number	Hits	Search Text	DB	Time stamp
1	1	crystallizing with (pzt or ferroelectric) with (Ar or argon) with oxygen	USPAT; US-PGPUB	2003/05/01 10:36
2	1	crystallizing with (pzt or ferroelectric) with (Ar or argon)	USPAT; US-PGPUB	2003/05/01 10:36
3	130	crystallizing with (pzt or ferrcelectric)	USPAT; US-PGPUB	2003/05/01 10:37
4	84	crystallizing with (pzt or ferroelectric) and oxygen and (ar or argon or nitrogen)	USPAT; US-PGPUB	2003/05/01 10:37
5	81	((crystallizing with (pzt or ferrcelectric)) and oxygen and (ar or argon or nitrogen)) not (crystallizing with (pzt or ferroelectric) with (Ar or argon))	USPAT; US-PGPUB	2003/05/01 10:37
6	73	((crystallizing with (pzt or ferrcelectric)) and (oxygen and (ar or argon or nitrogen)) not (crystallizing with (pzt or ferroelectric) with (Ar or argon))) and capacitor	USPAT; US-PGPUB	2003/05/01 11:30
7	3623	438,3,240,253,381,396.ccls.	USPAT; US-PGPUB	2003/05/01 11:47
8	786	438,3,240,253,381,396.ccls. and capacitor and ferroelectric	USPAT; US-PGPUB	2003/05/01 11:47
9	460	(438/3,240,253,381,396.ccls. and capacitor and ferrcelectric) and @adk=20000417	USPAT; US-PGPUB	2003/05/01 11:47
10	2194	257/295,303,306,310.ccls.	USPAT; US-PGPUB	2003/05/01 11:47
11	745	257/295,303,306,310.ccls. and capacitor and ferroelectric	USPAT; US-PGPUB	2003/05/01 11:47
12	476	(257/295,303,306,310.ccls. and capacitor and ferroelectric) and @adk=20000417	USPAT; US-PGPUB	2003/05/01 11:47

US-PAT-NO: 6201271

DOCUMENT-IDENTIFIER: US 6201271 B1

TITLE: Semiconductor memory device
prevented from deterioration
due to activated hydrogen

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As an electrode material of the ferroelectric capacitor, platinum has widely been used for both the upper electrode and the lower electrode for such a reason that it has a resistivity to a high-temperature oxidative atmosphere for crystallizing the ferroelectric material. As a ferroelectric material to be used for the ferroelectric capacitor, attention is focused on SrBi₂ Ta₂O₉ (SBT) and Bi₂Ti₃O₁₂ (BIT) that have excellent fatigue properties and are able to be operated at low voltage as compared with PbZr_xTi_{1-x}O₃ (PZT) having been examined to the detail, and the former substances are under elaborate examination.

According to an embodiment of the present invention, hydrogen sintering for the final recovery can be performed without deteriorating the ferroelectric characteristic nor the highly dielectric characteristic. Both the upper electrode and the ferroelectric material or the highly dielectric material are constructed of oxide materials, and therefore, the elements and oxygen atoms constituting the ferroelectric material or the highly dielectric material scarcely move even in the heat treatment in the subsequent production stage or aging during use, for which the fatigue characteristic of

the ferroelectric material and the reduction with time in dielectric constant of the highly dielectric material are also improved.

FIG. 4 is a sectional view of the plasma TEOS film of FIG. 3 improved in shape by argon sputtering;

Further, in order to improve the adherence of the formed lower electrode, annealing was effected for 30 minutes in nitrogen at a temperature of 600.degree. C. in an electric furnace.

Subsequently, the PZT film 11 was formed to a film thickness of 2000 .ANG. by the sol-gel method. The above PZT film forming method is performed by firstly dissolving lead acetate, titanium isopropoxide and zirconium isopropoxide into a solvent of 2-methoxyethanol so that the ratio of Pb:Ti:Zr=100:52:48 thereby forming a sol-gel material solution, coating this material solution on a silicon wafer formed to the lower electrode by means of a spinner rotating at a rotating speed of 3000 rpm, drying the wafer at a temperature of 150.degree. C. for 10 minutes in the atmospheric air and thereafter performing temporary baking at a temperature of 400.degree. C. for 30 minutes in the atmospheric air. The coating is repeated three to five times so that the desired film thickness of 2000 .ANG. is achieved. Subsequently, a heat treatment for crystallization is performed by the RTA (Rapid Thermal Annealing) method at a temperature of 650.degree. C. for 30 seconds in an atmosphere of a mixture of nitrogen and oxygen. In this stage, the flow rates of nitrogen and oxygen were set so that nitrogen flow rate: oxygen flow rate=4:1.

Next, the PZT film 11 and the lower electrode 10 were processed to a size of, for example, about $2.6 \text{ } \mu\text{m} \times 2.6 \text{ } \mu\text{m}$ and $3.0 \text{ } \mu\text{m} \times 3.0 \text{ } \mu\text{m}$ by the dry etching method, respectively. Subsequently, the titanium oxide film 12 was formed by the RF magnetron reactive sputtering method to a thickness of $250 \text{ } \text{Å}$, as a diffusion preventing layer of the elements constituting the ferroelectric capacitor. The ozone TEOS film 13 was formed as a second interlayer insulating film by the CVD method, and the titanium film 14 was further formed by the RF magnetron sputtering method to a thickness of $300 \text{ } \text{Å}$, as a layer for adhesion to the upper electrode of the capacitor to be formed on it. Next, a hole of, for example, about $1.8 \text{ } \mu\text{m} \times 1.8 \text{ } \mu\text{m}$ was formed by the dry etching method through the titanium film 14, the second interlayer insulating film 13 and the titanium oxide film 11 above the ferroelectric capacitor, so that a contact hole was opened up to the surface of the PZT film 11. The PZT film 11 was damaged by plasma exposure due to the dry etching. In order to recover this, a heat treatment was performed at a temperature of 500°C . for 30 seconds in oxygen by the RTA (Rapid Thermal Annealing) method.

The elemental composition ratio of the formed alloy oxide film 15 of platinum and rhodium was expressed by the ratio of platinum: rhodium oxygen= $70:15:15$. The film formation was performed by the RF magnetron reactive sputtering method while adjusting the total gas flow rate so that the reaction chamber comes to have a pressure of 10 mTorr at a gas flow rate of argon:oxygen= $2:1$. If the rhodium content increases with respect to the composition ratio of platinum to rhodium, then the

crystallinity of platinum having a peak of (111) becomes worse, and this is disadvantageous because a volumetric expansion will be significant during heat treatment in the subsequent process. Therefore, the rhodium content must be not greater than 20 percent in terms of the composition ratio thereof to platinum. Furthermore, unless a rhodium content of not smaller than 10 percent is achieved, no sufficient oxide results, so that the effect of platinum atoms for activating hydrogen cannot be suppressed. Therefore, rhodium must be not smaller than 10 percent and not greater than 20 percent in terms of the composition ratio thereof to platinum. Furthermore, for the same reason as above, the oxygen content relative to all the elements must be not smaller than 10 percent and not greater than 17 percent.

Next, the titanium nitride film 16, the alloy oxide film 15 of platinum and rhodium and the titanium film 14 were processed into a drive line shape by the dry etching method using chlorine gas. As stated before, the P3T film is also indirectly damaged by plasma exposure due to this dry etching. Therefore, for the purpose of recovering this, a heat treatment was performed at a temperature of 550.degree. C. for 30 seconds in oxygen by the RTA method.

In view of the above, the plasma TEOS film was firstly formed to a thickness of 3000 .ANG. as the third interlayer insulating film. Since an overhung shape (see the reference character D in FIG. 3) appeared even at this degree of film thickness, the shape was improved as shown in FIG. 4 by reverse sputtering using argon, and thereafter the ozone TEOS film was formed to a thickness of 7000 .ANG.. This film contained a certain amount of

moisture, and therefore, the plasma TEOS film was formed to a thickness of 3000 .ANG. for the prevention of the upward diffusion of moisture. The contact hole was formed in this place and the aluminum lead electrode 18 extending from the source/drain region was formed by the DC magnetron sputtering method.

Furthermore, a similar capacitor was formed of platinum for the capacitor upper electrode by the prior art technique, and the capacitor to which the present invention is applied and the prior art capacitor were concurrently subjected to a heat treatment for 10 minutes in an atmosphere of five percent of hydrogen and 95 percent of nitrogen while changing the heat treatment temperature, and the residual dielectric polarization values were measured after the heat treatment. As shown in FIG. 6, it was discovered that the residual dielectric polarization value of the capacitor using the prior art technique was reduced at and around the temperature of 180.degree. C. and the residual dielectric polarization value of the capacitor of the present invention was not reduced until the temperature of 430.degree. C.

FIG. 3 shows the second embodiment of the present invention, where the same components described with reference to FIG. 1 are denoted by the reference numerals and no description is provided for them. In FIG. 8 are shown a tungsten plug 19 formed for making contact of the source/drain region 5 with the capacitor lower electrode, an alloy film 20 of platinum and rhodium formed on the tungsten plug, an alloy oxide film 21 of platinum and rhodium formed as a diffusion barrier and oxygen preventing film, a platinum film 22 for improving the morphology of the ferroelectric film, and a

titanium oxide film 12 that serves as a diffusion preventing layer of elements of the ferroelectric film 23 formed on the lower electrode 22. In FIG. 8 are also shown a SrBi₂Ta₂O₉ (SBT) film 23 formed on the lower electrode 22 and an alloy film 24 of platinum or platinum and rhodium for a drive line interconnection.

Subsequently, tungsten is buried in the contact hole by the CVD method, and thereafter the surface is flattened by the CMP (Chemical Mechanical Polishing) method, thereby forming the tungsten plug 19. The alloy film 20 of platinum and rhodium was formed to a film thickness of 700 .ANG. on this tungsten plug 19 by the DC magnetron sputtering method, thereafter alloy oxide film 21 of platinum and rhodium to a film thickness of 300 .ANG. on the alloy film 20 by the DC magnetron reactive sputtering method, and the platinum film 22 was further formed to a film thickness of 500 .ANG. by the DC magnetron sputtering method. The elemental composition ratio of the formed alloy film of platinum and rhodium was expressed by the ratio of platinum:rhodium=80:20. The elemental composition ratio of the alloy oxide film of platinum and rhodium was expressed by the ratio of platinum:rhodium:oxygen=70:15:15. The film formation was performed by the magnetron reactive sputtering method while adjusting the total gas flow rate so that the reaction chamber come to have a pressure of 10 mTorr at a gas flow rate of argon:oxygen=1:1.

Next, a material solution of SrBi₂Ta₂O₉ (SBT) was coated on this lower electrode by a spinner rotating at a rotating speed of 3000 rpm by the MOD (Metal Organic Deposition) method and dried at a temperature of

250.degree. C. for five minutes. First baking was performed at a temperature of 600.degree. C. for five minutes in an oxygen atmosphere at the atmospheric pressure, and subsequently second baking, i.e. a heat treatment for crystallization, was performed in an oxygen atmosphere at a temperature of 50.degree. C. for five minutes by the RTA method. The processes from the coating to the heat treatment for crystallization were repeated three to five times so that an SBT film 23 came to have the desired film thickness of 2000 .ANG.. The forming method may be the sputtering method or the MC CVD (Metal Organic CVD) method besides the MOD method. In this case, when the composition ratio of rhodium increases in the alloy film of platinum and rhodium, the crystallinity of platinum having a peak of (111) and the film flatness become worse, so that the leak current characteristic of the ferroelectric film that lies in the upper layer deteriorates. Therefore, rhodium must have an element ratio of not greater than 80 percent relative to platinum. If the rhodium content is small in the alloy oxide film of platinum and rhodium, no sufficient oxide film results to reduce the oxygen blocking effect, so that the surface of the tungsten plug is oxidized during annealing in the presence of oxygen, consequently causing an electrical defective continuity. Therefore, rhodium must have an element ratio of not smaller than 10 percent relative to platinum in the alloy oxide film of platinum and rhodium.

Next, the titanium nitride film 16 and the film 24 that is alloy film of platinum and rhodium or platinum, and the titanium 14 were processed into a drive line shape by the dry etching method using chlorine gas. The SBT film is also indirectly damaged by plasma exposure due to this dry

etching. Therefore, for the purpose of recovering this and improving the leak characteristic of the ferroelectric capacitor, a heat treatment was performed in an oxygen atmosphere at a temperature of 800.degree. C. for 15 minutes.

Next, using a material of TEOS (tetraethylxysilane), the third interlayer insulating film 17 comprised of a plasma TEOS film of 3000 .ANG., an ozone TEOS film of 7000 .ANG. and a plasma TEOS film of 3000 .ANG. was formed. In general, the plasma TEOS film has a small moisture content in the film, however, it has a great amount of hydrogen content. The plasma TEOS film also tends to have a shape as indicated by the reference character D in FIG. 3, when its thickness is increased. On the other hand, the ozone TEOS film has a small amount of hydrogen content in the film, however, it has a great amount of moisture and a groundwork dependency. Consequently, the film thickness of the interlayer insulating film varies as shown in FIG. 2. In view of the above, the plasma TEOS film was firstly formed to a thickness of 3000 .ANG. as the third interlayer insulating film. Since an overhung shape (see the reference character D in FIG. 3) appeared even at this degree of film thickness, the shape was improved as shown in FIG. 4 by reverse sputtering using argon, and thereafter the ozone TEOS film was formed to a thickness of 7000 .ANG.. This film contained an increased amount of moisture, and therefore, the plasma TEOS film was formed to a thickness of 3000 .ANG. for the prevention of the upward diffusion of moisture. The contact hole was formed in this place and the aluminum lead electrode 18 extending from the source/drain region 5 was formed by the DC magnetron sputtering method.

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DOCUMENT-IDENTIFIER: US 6287637 B1
TITLE: Multi-layer approach for optimizing
ferroelectric film performance

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FIGS. 2(a), (b), (c), and (d) show the comparison of switched charge, V(90%), switched charge loss, and opposite state charge aging rate for the two types of PZT films, one of which is annealed in an oxygen ambient atmosphere, the other of which is annealed in a combined ambient atmosphere of argon and oxygen; and

In the preferred embodiment, a Gryphon Sputtering Deposition Tool is used. All layers 14, 16, and 18 are deposited at the same temperature (Room Temperature) and in the same sputter ambient environment (Argon). The sputter conditions for the nucleation layer 14 are 750 watts at an 18 mTorr pressure to produce a film between 50 and 100 Angstroms thick with approximately 30% excess lead. The bulk layer 16 is deposited at 1000 watts at a 2 mTorr pressure to build up the remaining film thickness with an excess lead content of about 15-20%. If an optional cap layer 18 is employed to enhance the top electrode interface characteristics, the bulk layer 16 thickness is reduced by the additional thickness of the cap layer 18. The cap layer 18 is between 200 and 300 Angstroms thick and varies between 25 and 35% excess lead. The sputter conditions for producing the cap layer 18 lead contents are

750 watts at a 10 mTorr pressure for 25% excess lead and 550 watts at an 18 mTorr pressure for 35% excess lead.

According to another method of the present invention, a specific anneal sequence is utilized after PZT deposition to control the crystalline texturing of the film, and by doing so, enhancing low voltage performance. In general, "texturing" is defined as preferred orientation and grain structure. This is accomplished by modifying the lead content in the PZT film to take advantage of specific phase formations that enhance PZT texturing. A multi-layer PZT is utilized to obtain a specific lead profile, with an associated anneal sequence to take advantage of that profile. The anneal sequences involve a two step process wherein a first step utilizes a low temperature argon ambient anneal to drive the formation of a platinum-lead and platinum-titanate intermetallic phases at the bottom electrode interface and a second step utilizes an oxygen anneal to complete the PZT crystallization process.

The multi-layer processing of the present invention is used because considerably more lead is required near the bottom electrode interface to supply the platinum-lead intermetallic layer; this high level of lead cannot be sustained in the bulk of the PZT film without an adverse impact on the kinetics driving crystal formation. The argon ambient anneal is used in the first step to prevent over saturation of the bottom electrode interface with oxygen. Excessive oxygen shuts down the desirable platinum-lead phase and favors an undesirable lead oxide (PbO) phase, which has a detrimental impact on PZT texture. By using the argon sequence of the present invention to establish the

platinum-lead and platinum titanate intermetallic phases, the energy required to produce a favorable texture during the oxygen anneal sequence can be reduced. Since the required energy is reduced, the stress/strain relationships in the lattice are reduced and the crystal forms along preferred orientations dictated by the nucleation foundation layer. Depending upon the PZT dopants used, the argon anneal generates a strong <math><111></math> orientation preference or a mixed texturing of <math><001></math> and <math><111></math> orientations. In particular, the quality of the <math><001></math> texturing directly influences the low voltage performance of the film. An additional benefit of reducing the energy requirements for the anneal is the conservation of lead in the bulk of the film. This conservation of lead enhances fatigue performance.

The argon and oxygen anneal sequence of the present invention can be modified by the addition of certain dopant materials to further enhance low voltage performance. A PZT film can be doped with lanthanum, calcium, and strontium dopants to achieve low voltage ferroelectric operation. Operation as low as three volts has been demonstrated, and the potential exists for operation at even lower voltages less than three volts. In addition, the argon and oxygen anneal described herein can be applied to other ferroelectric compositions as well as with other electrode structures and materials.

Utilizing the multi-layer PZT structure in conjunction with the two step anneal process enhances ferroelectric performance by establishing sufficient lead in the nucleation layer to adequately establish the intermetallic phases of lead platinum (Pb--Pt) and lead titanate (Pb--Ti).

These phases provide the foundation and seeding for proper PZT nucleation with orientation for optimum ferroelectric performance. The nucleation layer is kept thin (about 100 to 200 Angstroms) and lead rich (30%. \pm .5%) and provides the lead necessary to complete the phase interactions for the time and temperature window of the argon portion of the anneal. The argon anneal step is desirable to enhance the Pb--Pt and Pb--Ti interactions without the presence of oxygen. Typically, oxygen-only anneals overrun these interactions with the formation of PbO (lead oxide), which disrupts the preferred phase formations. The PbO formation is detrimental to preferred crystal orientation (<100>; and <110>; mixed orientations). After the argon portion of the anneal, the foundation is set for good crystal orientation, <111>; and <001>;, but the lattice remains oxygen deficient. The second anneal is done in an oxygen ambient atmosphere to complete the crystal formation, which is now driven by the foundation established with the argon anneal. The oxygen anneal fills the oxygen vacancies and completes the crystalline structure for the bulk of the film. The bulk of the PZT film can not be as lead rich (bulk layer lead =15%. \pm .5%) as the nucleation layer or excessive amounts of PbO would be generated during the oxygen anneal and again disrupt preferred crystal orientation in the bulk of the film.

Step 1 (Argon Anneal)

Argon ambient atmosphere;

Step 2 (Oxygen Anneal)

Oxygen ambient atmosphere;

After annealing, the wafer receives a top electrode deposition (1750 Angstroms of platinum), patterning and etch followed by a furnace anneal process at 650.degree. C. for 60 minutes in an oxygen ambient atmosphere, at which point the wafer is ready for electrical testing.

The main object of this embodiment of the present invention is the application of reduced O.₂ partial pressure ambient during PZT thin film crystallization. **Crystallizing PZT** thin films in reduced O.₂ partial pressure ambient results in better ferroelectric performance, cross wafer uniformity and wafer-to-wafer repeatability compared with pure O.₂ crystallization ambient.

Ferroelectric PZT [Pb(Zr,Ti)O.₃] thin film is one of the key components in FFAM.ETM. technology. After being deposited on a substrate, the PZT thin film needs to be annealed at elevated temperature, such as 600.degree. C., to form a polycrystalline thin film with a complex perovskite structure. Since PbO is very volatile at high temperature, say above 500.degree. C., the lead loss in the area close to the surface of the PZT film is higher than that in the bulk of the PZT film, resulting in a non-uniform distribution of lead in the direction of the thickness of the film. Adding excess lead in the PZT film before crystallization may compensate the lead loss in the surface area during high temperature anneal and form the perovskite phase. However, the lead content in the bulk of the PZT film may be too high. It may be difficult for lead to diffuse out of the film as the perovskite phase may block the migration of lead cations. Since the Pb--O bonding in PbO is much weaker than that in a perovskite structure, it is therefore more difficult for

lead cations to migrate through a perovskite phase. One way to increase the mobility of lead cations in a perovskite phase is to generate oxygen vacancies in the perovskite structure. It is known that the perovskite structure can tolerate vacancy concentration up to 20%. It is therefore possible to crystallize as-sputtered PZT film in a reduced O₂ partial pressure ambient so that the perovskite structure can still form but with a large amount of O₂ vacancies embedded in the structure. O₂ vacancies provide effective paths for lead cations to migrate in the film during high temperature sintering, giving rise to a more uniform lead distribution and more homogeneous formation of a perovskite phase.

There are two ways to generate a reduced O₂ partial pressure ambient. One is to use vacuum environment and flow a small amount of O₂ gas into the annealing chamber. Another way, according to the present invention, is to simultaneously flow O₂ and another type of gas that will not react with PZT film, such as argon, into the annealing chamber during high temperature crystallization. Method one requires an expensive pumping system and a good vacuum control system. Method two according to the present invention only requires mass flow controllers and a reasonable seal of the annealing chamber.

The assignee of the present invention, Ramtron International Corp. has used Argon rich ambient gas to crystallize PZT films for FRAM applications and parts containing PZT film annealed in an Argon/oxygen ambient atmosphere have been built and evaluated. The oxygen partial pressure in Ramtron's RTA (Rapid Thermal Anneal) chamber is in the range of 10.⁻⁴ to 100 Torr.

Experiments have been conducted to evaluate the ferroelectric performance of PZT films crystallized in the Argon rich ambient according to the present invention (Ar 0._{sub}.2 mixture followed by O._{sub}.2 anneal--herein Ar/O._{sub}.2 then O._{sub}.2), and for comparison, O._{sub}.2 only ambient followed by O._{sub}.2 anneal--herein O._{sub}.2 then O._{sub}.2. FIGS. 2(a), (b), (c), and (d) show the comparison of switched charge V(90%), switched charge loss, and opposite state charge aging rate for the two types of PZT films it can be seen that the PZT films crystallized in the combined Argon and O._{sub}.2 ambient of the present invention (i.e., Ar/O._{sub}.2 then O._{sub}.2) indeed show better ferroelectric performance compared with samples crystallized in the O._{sub}.2 ambient (i.e., O._{sub}.2 then O._{sub}.2) (that is, higher switch charge (Q._{sub}.SW), lower V._{sub}.90 %, lower Q._{sub}.SW loss @ 10._{sup}.9 cycles, and comparable opposite state switched charge aging rate).

FIG. 3 is a diagram of an RTA (Rapid Thermal Anneal) chamber for annealing a PZT wafer according to the present invention. An RTA chamber 32 includes heating coils 36 powered by a heating control unit 44. The RTA chamber 32 further includes a substrate or platform 38 for supporting a PZT wafer 40. A thermocouple 42 provides feedback to heating control unit 44. The RTA chamber 32 further includes a door 34 for introducing the wafer to the chamber. Annealing gasses are provided through oxygen cannister 46, nitrogen cannister 48, and oxygen cannister 50, which are introduced to the RTA chamber 32 under control of valves 52, 54, and 56, as required.

It is important to note that the combined argon and oxygen anneal of the

present invention can be used on either single layer, bi-layer, or tri-layer PZT films. Also, other inert gasses besides argon can be used, such as nitrogen or helium, or the like. The exact temperature and partial pressure of oxygen can be changed as required. A second anneal in oxygen can also be performed.

Having described and illustrated the principle of the invention in a preferred embodiment thereof, it is appreciated by those having skill in the art that the invention can be modified in arrangement and detail without departing from such principles. For example, if desired, the method of fabricating a ferroelectric multi-layer thin film according to the present invention can be modified to include only a single PZT layer, followed by the steps of annealing the single PZT layer in an argon ambient atmosphere, and then annealing the single PZT layer in an oxygen ambient atmosphere. Improvements in performance will still be obtained, even though a single PZT layer is used. We therefore claim all modifications and variations coming within the spirit and scope of the following claims.

annealing the PZT film in a combined argon and oxygen ambient atmosphere during PZT thin film crystallization; and

thereafter annealing the PZT film in an oxygen ambient atmosphere.

8. The method of claim 7 in which the second gas is argon.

9. The method of claim 7 in which the second gas is nitrogen.

12. The method of claim 7 in which the oxygen partial pressure in the

annealing chamber during said high temperature
crystallizaton is in the range
of 10.sup.-4 to 100 Torr.